

Anomalous Magnetic Susceptibility in Iron Pnictides: Paramagnetic Phase

M.S. Laad and L. Craco

Max-Planck-Institut für Physik komplexer Systeme, 01187 Dresden, Germany

(Dated: March 22, 2009)

Observation of an anomalous temperature dependence of the spin susceptibility, along with a spin gap in NMR, in the quantum paramagnetic *normal* state of Iron Pnictides is a signature of an unusual metallic state. We argue that *both* these anomalous features are associated with a wide fluctuational regime dominated by dynamical, short-ranged and frustrated spin correlations in a strongly correlated metal. Using LDA+DMFT, we show that *both* these features can be quantitatively understood in the doped Fe-pnictides. We argue that such spin correlations naturally arise in a *Mottness* scenario, where an *effective*, dualistic description involves coexisting renormalized quasiparticles and effectively localized moments, arising from the same set of *d*-bands.

PACS numbers: 74.70.-b, 74.25.Ha, 71.27.+a

The precise mechanism of unconventional superconductivity (U-SC) in the recently discovered Iron Pnictides (FePn) is presently a hotly debated issue.¹ U-SC arises on the border of a geometric frustration (GF)-induced “stripe”-spin-density-wave (SSDW).² Short-ranged, frustrated spin correlations, expected to survive doping-induced destruction of SSDW order,³ are then expected to play an important role in the normal and SC phases of FePn.

Study of magnetic fluctuations in an U-SC can help unearth the symmetry of the SC order parameter, as shown by detailed studies for cuprates⁴. In FePn, extant experiments already show anomalous behavior. Specifically, NMR studies reveal normal state pseudogap behavior⁵ and U-SC, as seen from opening up of a spin gap at $T^* = 150 - 200$ K, and $T_1^{-1} \simeq T^n$ with $n = 2.2 - 2.5$ for $T \ll T_c$. However, other probes reveal anisotropic, albeit fully gapped, structure of the in-plane gap function. While this is the expected response of a GF spin system in its quantum disordered phase,² FePn are *metals*, albeit presumably close to a Mott-Hubbard instability.² Are there other signatures of anomalous magnetic correlations in FePn? Extant data reply in the affirmative: the *uniform* spin susceptibility shows a *linear-in- T* dependence for $T > T_{SSDW}(x)$ up to 800 K for *both* the 1111- and 122-FePn.^{6,7} This anomalous behavior persists upon destruction of the SSDW, even going over to a $\chi(T) \simeq T^{1+n}$ form at lower T , with $n \leq 1$. Actually, just above T_c for the doped La-based 1111-FePn with $x > 0.05$, the spin susceptibility obeys the $\chi(T) = \chi_0 + AT^{1+n}$ law, and the linear-in- T dependence is recovered smoothly only at higher T . This observation is strong circumstantial evidence for relevance of GF, since the $\chi(T) \simeq T$ law is also seen above T_N in another GF system, $Na_{0.5}CoO_2$, which is also a poor metal like doped FePn.⁸ However, *Cr* and its alloys, widely believed to be described in a more itinerant framework, also show the linear-in- T susceptibility. The distinguishing feature of the FePn with $x \simeq 0.1$ is, however, that they exhibit the spin gap in NMR in the quantum *disordered* phase. This observation puts them into the “strong coupling” category, since the spin gap in the paramagnetic phase

is beyond reach of an itinerant description. A consistent theory of magnetic fluctuations in FePn *must* then reconcile the spin-gap in NMR with $\chi(T) \simeq T^{1+n}$, with $0 \leq n \leq 1$ in a single picture.

Earlier theoretical studies have focussed on both itinerant weak- and strong coupling routes.^{6,9} In the itinerant view, a $\chi(T) \simeq T$ *requires* nesting of the electron- and hole-like Fermi sheets. This involves fine tuning, since no such nesting can be invoked for doped 1111-FePn, even as the $\chi(T) \simeq T$ behavior persists. The second view, based on (presumably) Kondo-like coupling of itinerant carriers to effectively *localized* spins in a $J_1 - J_2$ -Heisenberg model, does not require fine tuning, but is strictly valid only in the localized regime.² Experimental evidence, however, places the FePn in the intermediate-to-strong correlation regime,² where the *dualistic* aspect of self-consistently correlated *d* electrons should manifest itself. In this *Mottness* scenario, *iboth* the “itinerant” carriers and the “local” moments arise from the same set of *d*-electrons, simultaneously giving renormalized LDA-like “bands” as seen in dHvA¹⁰ and “localized” magnetic responses seen in NMR and INS studies. We thus use LDA+DMFT, which is known to capture “Mottness” *exactly* at the mean-field level.^{11,12}

In a study of magnetic fluctuations, the central quantity of interest is the *dynamical* spin susceptibility, $\chi(\mathbf{q}, \omega) = \sum_{a,b} \chi_{a,b}(\mathbf{q}, \omega)$, where *a, b* are *all d*-orbital indices, and \mathbf{q}, ω are the momentum and energy transfers in INS. Viewing FePn as strongly correlated systems, we construct $\chi(\mathbf{q}, \omega)$ in terms of the *full* LDA+DMFT propagators computed in earlier work.^{13,14,15} Good quantitative agreement between LDA+DMFT and key experiments in *both*, the normal and U-SC states, has been shown there, supporting our choice. The prescription is to replace the band Green functions used in weak-coupling RPA-like approaches¹⁴ by their LDA+DMFT counterparts. This ensures that the dynamical aspect of strong, local, multi-orbital (MO) correlations is included from the outset.

For the incoherent “normal” metallic state in FePn, after replacing the *bare* $G_{aa}(\mathbf{k}, \omega)$ with $G_{aa}(\mathbf{k}, \omega) \equiv G_{aa}^{LDA+DMFT}(\mathbf{k}, \omega) = [\omega - \epsilon_{ka} - \Sigma_a(\omega)]^{-1}$, and intro-

ducing the spin operator $S_{a,\mu}(\mathbf{q}) = \frac{1}{2} \sum_{\mathbf{k}} c_{a,\mu,\sigma}^\dagger(\mathbf{k} + \mathbf{q}) \sigma_{a,\sigma,\sigma'}^\mu c_{a,\mu,\sigma'}(\mathbf{k})$, with $\mu = x, y, z$, the “bare” dynamical spin susceptibility reads

$$\chi_{0,a,b}^{\mu\nu}(\mathbf{q}, \omega) = K_{\mu\nu}^{\sigma\sigma'} \int d\nu \int d\epsilon \sum_{\mathbf{k}, \omega'} \rho_{aa\sigma}(\mathbf{k} + \mathbf{q}, \nu) \rho_{bb\sigma'}(\mathbf{k}, \epsilon) \frac{n_F(\nu) - n_F(\epsilon)}{\omega + \nu - \epsilon + i\eta},$$

where $K_{\mu\nu}^{\sigma\sigma'} \equiv \frac{1}{2} \sigma_{a,\sigma\sigma'}^\mu \cdot \sigma_{b,\sigma\sigma'}^\nu$, the σ are Pauli matrices, and n_F the Fermi function. $\rho_\alpha(\mathbf{k}, \nu) = (-1/\pi) \text{Im} G_{\alpha\alpha}(\mathbf{k}, \nu)$ is the one-particle spectral function for orbital α . Including the ladder vertex in an infinite summation of “ladder” diagrams using RPA, the renormalized magnetic susceptibility, $\chi_{a,b}(\mathbf{q}, \omega) = [\chi_{0,a,b}^{-1}(\mathbf{q}, \omega) - J(\mathbf{q})]^{-1}$, where, following¹⁶ $\chi_{0,a,b}(\mathbf{q}, \omega)$ is evaluated in DMFT. The “bare” bubble contribution is found as $\chi^{(0)}(0, \omega) \simeq \chi^{(0)}(\omega) = C \sum_{a,b} \int d\nu \rho_a(\nu) \rho_b(\omega + \nu) [f(\nu) - f(\omega + \nu)]$, and $J(\mathbf{q}) = J_1(\cos(q_x a) + \cos(q_y a)) + J_2 \cos(q_x a) \cos(q_y a)$, with $J_1 \simeq \frac{t_{ab}^2}{U' + J_H}$ and $J_2 \simeq \frac{t'_{ab}{}^2}{U' + J_H}$ being the frustrated superexchange scales in FePn.² Using $\chi_{0,a,b}(\mathbf{q}, \omega)$, the dynamical spin susceptibility, $\chi''(\mathbf{q}, \omega)$, can be now expressed in terms of the *full* DMFT propagators computed earlier for the incoherent “normal” state.¹³

Using $\chi(\mathbf{q}, \omega)$ at low T , the uniform ($\mathbf{q} = 0$) spin susceptibility is now estimated as

$$\chi(T) \equiv \chi(0, T) = \int_{-\infty}^{+\infty} \frac{\chi''(0, \omega) d\omega}{1 - e^{-\beta\omega}}.$$

Notice how frustration appears in the “bare” bubble comprised of fully renormalized DMFT propagators, which contain contributions from the (frustrated) hoppings (via DMFT propagators), as well as in $J(\mathbf{q})$ via the RPA sum. In a companion work,¹⁴ we have shown how this yields nice agreement with the NMR relaxation rate over the *full* T range for FePn,¹⁴ including spin gap formation and unconventional power-law form of T_1^{-1} below T_c . Here, we extend that study to investigate the spin susceptibility in detail. We explicitly demonstrate that $\chi(T) \simeq T$ at high T , with a smooth crossover to a $\chi(T) \simeq T^{1+n}$ form at lower T with n smoothly increasing from zero to unity as T is lowered, for the doped FePn which become superconducting at lower T . We show how *both* NMR and susceptibility data in the “normal” state can be rationalized in terms of strong, frustrated spin correlations arising in a multi-band, correlated system on the verge of *Mottness*.

Before presenting our results, we make a few relevant remarks. In contrast to earlier studies,^{6,9} our model incorporates *all* five d bands, and explicitly captures the anisotropic three-dimensional band structure of the 1111-FePn. This should become relevant at low T , and a *smooth* crossover to $D = 2$ physics should occur with increasing T above $T_{SDW}(x)$. In such a $D = 2$ regime, one

might associate the $\chi(T) \simeq T$ law with the fluctuational regime that occurs *below* the mean-field ($T_{MF} \simeq J_2 \gg T_{SDW}, T_{SC}$) crossover.⁶ The wide T range over which these latter exist points to dominant effects of non-local spatial spin correlations up to rather high T . Combining this with the GF nature of FePn *and* observation of the spin gap below T^* in the 1111-FePn suggests that the spatial extent of these spin correlations should be of order a $Fe - Fe$ unit cell. These are precisely the correlations accessed by our formulation above, though our neglect of vertex corrections of the “crossing diagram” type is an approximation. We note, however, that a similar approximation is shown to reconcile the ARPES lineshapes *and* the neutron scattering results in cuprates.¹⁷ This indicates that such vertex corrections are small, justifying our approximation.

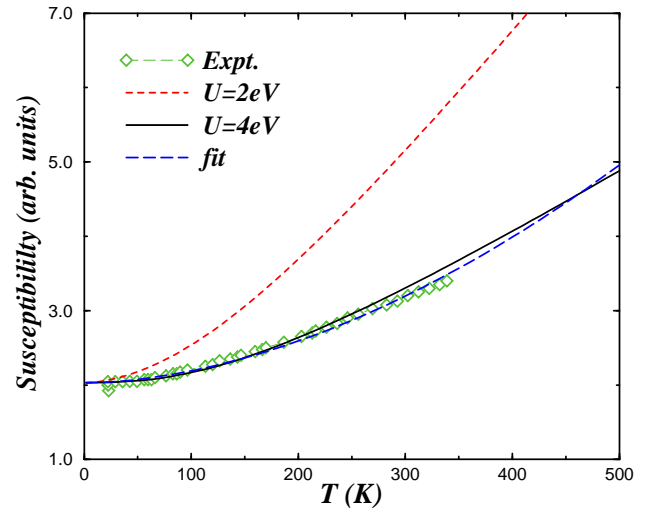


FIG. 1: (Color online) T -dependence of the uniform spin susceptibility, $\chi(T)$, for the $x = 0.1$ La-based Fe-pnictide. Notice the *smooth* evolution of the linear-in- T law at “high”- $T > 200$ K to a $T^{1.8}$ law at lower T , in nice agreement with experiment.⁷ Also note that the U -dependence of the susceptibility is strong evidence for a sizably correlated multi-orbital scenario in FePn. For clarity, both theoretical curves are shifted downward to coincide with the experimental curve at T_c .

We now present our results. In Fig. 1, we show the T -dependent uniform spin susceptibility, $\chi(T)$, for $LaO_{1-x}FeAsF_x$ at a representative electron doping, $x = 0.1$, for two values of U corresponding to intermediate-to-strong ($U = 4.0$ eV) and to weak ($U = 2.0$ eV) coupling. Since we do not treat the SSDW instability here, we focus only on the regime $x > 0.04$, where we study the T -dependent $\chi(T)$ up to lower T in the “normal” state. For $x = 0.1$, SSDW order is destroyed by doping. However, in full agreement with experiment, $\chi(T)$ continues to show the linear-in- T dependence in the range $200 \text{ K} < T < 800 \text{ K}$ (only shown up to 350 K), even exhibiting a smooth *increase* of curvature ($\simeq T^{1.8}$) at even

lower T in the vicinity of T_c . In fact, for $x = 0.1$, we have fitted $\chi(T)$ up to 500 K with the $2.03 + 0.42T^{1.8}$ law, quite distinct from the linear-in- T behavior seen for small x .⁷) Only at high T is the linear-in- T behavior recovered for $x \simeq 0.1$. The T -dependence of our computed susceptibility is very close to this fit to experiment, as shown in Fig. 1. Clearly, $\chi(T)$ with $U = 2.0$ eV substantially deviates from experiment, while a much better agreement is obtained with $U = 4.0$ eV, testifying to the importance of sizable multi-orbital electronic correlations in FePn. For sake of discussion, we show our earlier theoretical result for the NMR relaxation rate, $[T_1T]^{-1}$,¹⁴ with $U = 4.0$ eV in Fig. 2. The absence of a Korringa form ($[T_1T]^{-1} = \text{const}$) at any T is striking. Around $T^* \simeq 150$ K, clear, gradual opening up of a spin gap, again as seen in NMR work, is found. The correlation between the spin-gap in NMR and $\chi(T) \simeq T$ is not easily obvious: $\chi(T)$ continues to vary *smoothly* even as $[T_1T]^{-1}$ shows the spin gap behavior.

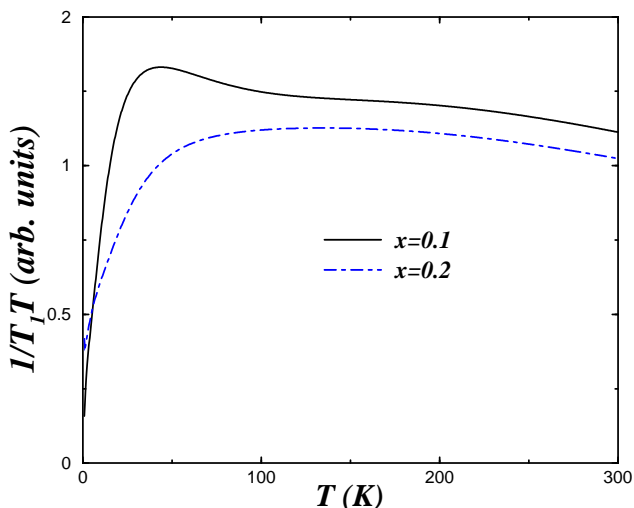


FIG. 2: (Color online) Low T behavior of the NMR $[T_1T]^{-1}$ for $\text{LaO}_{1-x}\text{FeAsF}_x$ with $x = 0.1, 0.2$, corresponding to doped samples with no SSDW order. The emergence of the spin gap around $T^* \simeq 150 - 200$ K for the doped FePn is clearly resolved, in good agreement with experiment.⁴

Such semi-quantitative agreement, even in details, with *both* NMR and spin susceptibility experiments is a very gratifying feature of our modelling. Deeper insight into these anomalous responses, and of their connection to U-SC in doped FePn, is clearly desirable. To this end, we start by noticing that $\chi(T) \simeq T$ is found in the range $0 < T < T_{MF} \simeq O(J)$ even for a classical, *unfrustrated* lattice in $D = 2$.¹⁸ Since the $D = 2$ Heisenberg model cannot have a finite T_N , it follows that this linear behavior must be associated with the fluctuational regime below the mean-field crossover, as discussed by Zhang *et al.* in the localized limit of the $J_1 - J_2$ model.⁶ In a GF system like FePn, this regime involves short-ranged, *dynamical* spin correlations above T_{SDW} . In “bad metal-

lic” FePn, the “Mottness” philosophy allows us to describe the low-energy magnetic fluctuations by an *effective* $J_1 - J_2$ model.² In this model, there is a wide T range, $T_{SDW} < T < T_s$, where translational symmetry is spontaneously broken but spin-rotational $\text{SU}(2)$ invariance is not.² Such a transition has interesting consequences:

(i) it naturally accounts for opening up of a spin gap, leading to consistency with NMR data, and,

(ii) it breaks the symmetry between $\mathbf{q} = (\pi, 0)$ and $(0, \pi)$, already above T_{SDW} . This breaks the lattice *rotational* symmetry (Ref. 3) and should lead to the tetragonal-orthorhombic (T-O) distortion,¹⁹ accompanied by a striped-SDW long range order at T_{SDW} . In fact, in general, $T_s > T_{SDW}$,¹⁹ supporting such a view. This could also lead to a nematic instability, a possibility that has already been proposed in the FePn context.²⁰

(iii) Finally, these dynamical *short-ranged* spin correlations should survive the doping-induced melting of the SSDW. These nearest- and diagonal neighbor correlations are attractive candidates for the “glue” that leads to short-coherence length U-SC in FePn. In fact, the U-SC gap function should then have nearest neighbor (n.n) and diagonal components: these *exactly* correspond to the ex- s and s_{xy} -pair components. Interestingly, rigorous analysis *requires* that *both* co-exist in FePn.²¹ This is precisely the in-plane, *nodeless* part of the SC gap function that we have proposed in Ref. 15 as an instability of the incoherent “normal” state in doped FePn. Since the pair wavefunction has only n.n and next-n.n components as above, this also affords a way to rationalize the short SC coherence length, high upper critical fields, and Uemura scaling, all of which are observed in FePn.²²

Hence, in our work, the anomalous spin susceptibility arises from effectively localized, short-ranged and sizably frustrated spin correlations above T_{SDW} in the 1111-FePn. That the FePn are bad metals (the drop in $\rho(T)$ at low T in some of the FePn, especially in $\text{LaO}_{1-x}\text{FeAsF}_x$, actually *correlates* with the opening of the spin gap,²³ and, cannot be taken as evidence of FL behavior)⁶ means that these short-ranged spin correlations are further damped by the dynamical charge fluctuations in reality. As a further check of our analysis, we notice that, choosing the spin gap, $\Delta \simeq 150 - 200$ K, from our NMR result, the *dc* resistivity is expressed as $\rho_{dc}(T) \simeq \rho_0 + CTe^{-\Delta/k_B T}$, in good agreement with the observations for $x = 0.1$ in LaOFeAs ;²³ interestingly, extraction of Δ from the *dc* resistivity gives $\Delta = 164$ K for $x = 0.1$, in good agreement with our estimate from NMR. When the spin gap closes, the *dc* resistivity recovers its characteristic linear-in- T behavior, as indeed seen.²³ This is again reminiscent of the evolution of $\rho_{dc}(T)$ in underdoped cuprates with increasing hole doping.⁴ In a “Mottness” scenario, with relatively stable local moments (this should not be confused with small sublattice *magnetization* in FePn, which can be drastically lowered by frustration),² the *effective* Heisenberg limit should be qualitatively correct below an exchange scale renormalized *downward* by metallicity. We empha-

size that this argument does not constitute a strict mapping from our DMFT results to an effective frustrated Kondo-Heisenberg model (for a more formal mapping, see Ref. 2); however, the latter does allow deeper insight into the DMFT results.

Finally, at lower T for $x \simeq 0.1$, the dimensional crossover from $D = 2$ to $D = 3$ should occur. This is what leads to the deviation of the low T susceptibility from the linear-in- T behavior. Our finding of an approximate $\chi(T) \simeq T^{1.8}$ law in this regime (in nice agreement with the $x = 0.1$ La-based 1111-FePn) suggests multi-orbital effects at work: in particular, we argue that small inter-layer couplings (hopping between d_{z^2} orbitals on neighboring layers, and concomitant superexchange, J_{\perp}), setting in at low temperatures, lead to anisotropic $D = 3$ physics, giving an approximate $\chi(T) \simeq T^2$ law, smoothly evolving from the linear-in- T variation at higher T .

Our analysis thus leads to a very different picture compared to the one obtained from an itinerant view.⁹ In the itinerant view, spin gap generation requires rather involved extensions of the HF-RPA approach hitherto used therein,²⁴ and these should bring the theoretical description closer to the limit considered here. Also, evolution of $\chi(T) \simeq T$ to the $T^{1.8}$ dependence with doping (for $x = 0.1$ and beyond see Ref. 6) remains to be clarified there. In our “strong coupling” picture, this behavior arises from the $2D - 3D$ crossover which should set in with increasing x at low T in the quantum paramagnetic metal. Using LDA+DMFT, we show how *both*, the $\chi(T) \simeq T$ at high T , as well as its T^2 -like behavior for low T in the $x > 0.06$ regime, can be obtained in a strongly

correlated picture. Both the linear-in- T susceptibility *and* the spin gap are thus associated with the wide ranged (in T) fluctuational contributions associated with short-ranged, geometrically frustrated spin correlations in the quantum disordered regime of an *effective*, doped $J_1 - J_2$ antiferromagnet. Our view is also fully consistent with ARPES,²⁵ optical²⁶ and μ SR²⁷ data, all of which are consistent with a strongly correlated metal giving way to U-SC.

In conclusion, we have studied the temperature dependence of the uniform spin susceptibility, $\chi(T)$, for the 1111-FePn using the first-principles LDA+DMFT method. Good semiquantitative agreement with published experimental data testifies to the relevance of short-ranged and dynamical multi-band electronic correlations in FePn. The same approach also yields a satisfying description of the NMR $[T_1T]^{-1}$ as a function of T in an earlier work.¹⁵ We thus identify *both*, the spin gap in NMR *and* T dependence of $\chi(T)$, as manifestations of the wide (in T) fluctuational regime of a sizably frustrated magnet between its actual ordering scale (T_{SDW}) and the mean-field crossover (T_{MF}). Our study pinpoints the crucial roles of strong multi-orbital electronic correlations and sizable geometric frustration to understand magnetic fluctuations in the incoherent metallic state of the 1111-Iron Pnictides.

Acknowledgments

The Authors thank the MPIPKS, Dresden, for hospitality and financial support.

-
- ¹ I.I. Mazin and J. Schmalian, arXiv:0901.4790.
 - ² Q. Si and E. Abrahams, Phys. Rev. Lett. **101**, 076401 (2008); ibid. J. Wu, P. Phillips, and A.H. Castro Neto, Phys. Rev. Lett. **101**, 126401 (2008); G. Baskaran, J. Phys. Soc. Jpn. **77**, 113713 (2008); Q. Si, E. Abrahams, J. Dai, and J.-X. Zhu, arXiv:0901.4112.
 - ³ C. Xu, M. Müller, and S. Sachdev, Phys. Rev. B **78**, 020501(R) (2008).
 - ⁴ H. He *et al.*, Phys. Rev. Lett. **86**, 1610 (2001); ibid. J. Bobroff *et al.*, Phys. Rev. Lett. **78**, 3757 (1997).
 - ⁵ Y. Nakai, S. Kitagawa, K. Ishida, Y. Kamihara, M. Hirano, and H. Hosono, arXiv: 0810.3569.
 - ⁶ G.-M. Zhang, Y.-H. Su, Z.-Y. Lu, Z.-Y. Weng, D.-H. Lee, and T. Xiang, arXiv:0809.3874.
 - ⁷ R. Klingeler, N. Leps, I. Hellmann, A. Popa, C. Hess, A. Kondrat, J. Hamann-Borrero, G. Behr, V. Kataev, and B. Büchner, arXiv:0808.0708.
 - ⁸ M.L. Foo, Y. Wang, S. Watauchi, H.W. Zandbergen, T. He, R.J. Cava, and N.P. Ong, Phys. Rev. Lett. **92**, 247001.
 - ⁹ M.M. Korshunov, I. Eremin, D.V. Efremov, D.L. Maslov, and A.V. Chubukov, arXiv:0901.0238.
 - ¹⁰ A.I. Coldea *et al.*, Phys. Rev. Lett. **101**, 216402 (2008).
 - ¹¹ G. Kotliar, S.Y. Savrasov, K. Haule, V.S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. **78**, 865 (2006).
 - ¹² K. Haule, J. H. Shim, and G. Kotliar, Phys. Rev. Lett. **100**, 226402 (2008).
 - ¹³ L. Craco, M. S. Laad, S. Leoni, and H. Rosner, Phys. Rev. B **78**, 134511 (2008); ibid M.S. Laad, L. Craco, S. Leoni, and H. Rosner, Phys. Rev. B **79**, 024515 (2009).
 - ¹⁴ L. Craco and M.S. Laad, arXiv:0903.1568.
 - ¹⁵ M.S. Laad and L. Craco, arXiv:0902.3400.
 - ¹⁶ A. Georges, G. Kotliar, W. Krauth and M. Rozenberg, Revs. Mod. Phys. **68**, 13 (1996).
 - ¹⁷ U. Chatterjee *et al.*, Phys. Rev. B **75**, 172504 (2007).
 - ¹⁸ D. Hinzke, U. Nowak, and D.A. Garanin, Eur. Phys. J. B **16**, 435 (2000).
 - ¹⁹ C. de la Cruz *et al.*, Nature **453**, 899 (2008).
 - ²⁰ S.A. Kivelson and H. Yao, Nature Materials **7**, 927 (2008).
 - ²¹ W.-L. You, S.-J. Gu, G.-S. Tian, and H.-Q. Lin, arXiv:0807.1493.
 - ²² Y.J. Uemura, arXiv:0811.1546.
 - ²³ G. Fuchs *et al.*, arXiv:0902.3498.
 - ²⁴ P. Monthoux and G.G. Lonzarich, Phys. Rev. B **66**, 224504 (2002).
 - ²⁵ L. Wray *et al.*, Phys. Rev. B **78**, 184508 (2008).
 - ²⁶ A.V. Boris *et al.*, Phys. Rev. Lett. **102**, 027001 (2009).
 - ²⁷ R. Prozorov *et al.*, arXiv:0901.3698.